ARKIV FÖR FYSIK Band 12 nr 13

Communicated 10 April 1957 by MANNE SIEGBAHN and ERIC HULTHÉN

On the systematics of transition probabilities of $M4$ transitions of the type $h_{11/2} \rightarrow d_{3/2}$

Decay of Te^{133m}

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With 4 figures in the text

ABSTRACT

A careful analysis is made of the radial matrix elements of $M4$ transitions of the type $h_{11/2} \rightarrow d_{3/2}$. It has previously been claimed that the matrix elements systematically increase towards the magic numbers. Especially the matrix element of Te^{133m} was found to be large in comparison with the matrix elements of the other Te isomers. For this reason the decay of Te^{133m} has been studied. The half-life of the isomeric state was found to be 53 ± 4 m with a decay energy of 334.0 ± 0.3 keV. Further, a large β-branching of 87 % from the isomeric level was found. This reduces the empirical matrix element of Te^{133m} to the order of magnitude of the other matrix elements in the isomers of tellurium. It must therefore be concluded that there are no certain experimental proofs that the matrix elements of $M4$ transitions are larger when shells are almost filled.

Introduction

The theoretically predicted life-time of a γ-transition of a nuclear state is known to be a function of the multipolarity, energy of the transition and the matrix element, the latter depending on the nuclear model adopted [1, 2]. A comparison of the matrix element obtained from experimental half-lives and energies of isomers with those calculated on the basis of the shell model, generally shows considerable deviations, except for $M4$ transitions. These occur in the three "islands of isomers", characterized by transitions between the one particle levels $g_{9/2} \rightarrow p_{\frac{1}{2}}$, $h_{11/2} \rightarrow d_{3/2}$ and $i_{13/2} \rightarrow f_{5/2}$, resp.[1] The empirical matrix elements of these $M4$- transitions are usually only a factor of 2 to 3 smaller than those predicted from the shell model [2].

Instead of looking at the absolute value of the matrix element, it is sometimes more interesting to compare its relative magnitude in isomers of a given element. In $M4$ transitions, which show the most regularities, several authors [2, 3, 4] have pointed out that the empirical matrix element increases close to a magic number. The explanation given was that, when approaching a closed shell, the last bound nucleon shows purer single particle transition properties, thus giving a larger matrix element.

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[1] It is usual to distinguish between proton and neutron isomers by setting P or N in front of the level nomenclature, together with a number indicating in which shell the transition-nucleon is situated.

Recently, however, it has been found [5] that in the odd Pb isomers ($i_{13/2} \rightarrow f_{5/2}$ transitions), the empirical matrix elements are remarkably constant within the experimental errors. The question arises whether there is a real effect in the "second island of isomers" (5 N $h_{11/2} \rightarrow d_{3/2}$ transitions) or whether the effect seen [2] depends on inaccurate measurements. For that reason the following aspects will be treated.

1. The experimental matrix elements will be calculated together with the uncertainty. It will be checked whether the small tendency, which is pronounced for Xe and Ba isomers, still exists when the experimental errors are taken into account.
2. Among the isomers of tellurium, a large deviation is known to exist in Te^{133m}. This will be investigated from an experimental point of view, giving special emphasis to a search for β-branching from the isomeric level.

Systematics of the empirical matrix elements of 5 N $h_{11/2} \rightarrow d_{3/2}$ transitions

Using a central nuclear potential, the radial matrix element (M) for an $M4$ transition between two states i and f may be written according to [2] explicitly as

$$M^2 = 1.65 \times 10^6 \times r_0^{-6} A^{-2} E_\gamma^{-9} S^{-1} \tau_\gamma^{-1}, \tag{1}$$

where r_0 = 1.20 [6]
A = mass number
E_γ = energy of the $M4$ γ-ray (in MeV)
S = statistical factor
τ_γ = mean life of the γ-ray.

The radial matrix element has been chosen so that a comparison with $M4$ transitions other than those of $h_{11/2} \rightarrow d_{3/2}$ can be made.

In the present case, the value of S is $\frac{15}{11}$ [2]. The τ_γ can be expressed in terms of the observed half life ($T_{1/2}$), conversion coefficients (α_K, α_L, ...) and β-branching (β)

$$\tau_\gamma = \frac{100\, T_{\frac{1}{2}}}{\ln 2\,(100-\beta)} \left\{ 1 + \alpha_K \left(1 + \frac{\alpha_L + \alpha_M + \dots}{\alpha_K} \right) \right\}.$$

In Table 1 values from the literature for energies, half-lives and β-branching of the isomers in the $h_{11/2} \rightarrow d_{3/2}$ region are given. Further, the empirical value of M^2 of the transition is calculated from (1). When no reference is given in the table, the value is taken from [7, 8]. The K-conversion coefficients are the theoretical values of L. Sliv *et al.* [9], which, for the elements treated here, differ from Rose's [10] with a few per cent only. For the ratio $(\alpha_L + \alpha_M + \dots)/\alpha_K$ the approximate expression $\frac{4}{3} \frac{\alpha_L}{\alpha_K}$ has been used.

When only one literature value exists, the uncertainty of the measurements are those given by the authors, where several values exist, a mean value has been preferred together with the maximum deviation. For the K/L ratio, an error of about (10–15)% is used. However, since M^2 is not usually very sensitive to small variation in K/L, it is not necessary to know this value more accurately. The uncertainty in the value of M^2 is the maximum error.

In Fig. 1 M^2 has been plotted as a function of the neutron number, together with the experimental errors. One notices first that the magnitude of the M^2 value is about a factor of three larger than those reported earlier [2, 17]. This is due to the

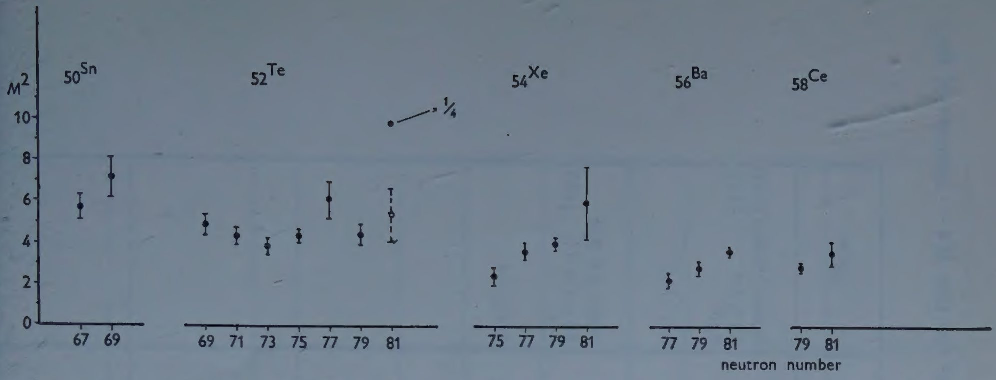

Fig. 1. The empirical value of M^2 as a function of the neutron number, together with the uncertainty of the M^2 value for the isomers of Sn, Te, Xe, Ba and Ce. The point with a dashed line indicating its error is the value obtained in this investigation.

fact that a smaller radius has been used here in (1) [ref. 6]. It is further seen that for Xe and Ba a small tendency for the empirical matrix element to increase close to a filled shell exists, even when the uncertainty of the measurements is indicated. There appears also to be a small increase of the value of M^2 when approaching the proton number 50. The only case showing a large deviation is in Te^{133m}. The energy used to calculate the value of M^2 for this isomer had been measured with low resolution so that the error in the matrix element is larger than 100%. However, with the more accurate energy determination made in this investigation, the matrix element was still quite outside the domain of the other matrix elements of the Te isomers. The tendency for the value of M^2 of the Te isomers to increase with neutron number was more pronounced when this discussion first started. The value of M^2 of Te^{131m} was then ~ 8. Recently, however, E. Hebb [11] has found a larger β-branching in this isomer than was reported earlier [21], thus reducing the value of M^2 to 4.6. It is possible that the large deviation of the value of M^2 for Te^{133m} will also be reduced by a β-branching not previously observed.

It is interesting here to compare the matrix element given above with those of the isomeric groups 6 N $i_{13/2} \rightarrow f_{5/2}$ and 5 P $h_{11/2} \rightarrow d_{3/2}$. The first group consists of the Pb isomers. As has been mentioned, the M^2 value of these appear to be constant (within the experimental errors) and a mean value of $M^2 = 3.7$ is obtained [5]. These thus show a slightly smaller value than the mean value of M^2 of the isomers 5 N $h_{11/2} \rightarrow d_{3/2}$. In the second group, only two isomers are known, Au^{197m} and Au^{195m} [16]. With the experimental data given in [16] one obtains for M^2 1.9 and 3.5 resp. The one-particle model predicts a value of M^2 for proton isomers twice that of neutron isomers [2]. However, the experimental matrix elements of the $M4$ transitions in the 4 N and 4 P groups show no such difference [2]. The same appears to be true for the 5 N and 5 P group.

Experimental procedures and results of the investigation of Te^{133m}

Te^{133m} was produced in the fission of uranium by slow-neutron bombardment. After the chemical separation, the energy and half-life of the isomeric transition was measured in a double focusing spectrometer. The β-branching was in principle measured from the known yield in fission and a comparison with Te^{131m}.

Table 1. Energy (E_γ), half-life ($T_{\frac{1}{2}}$), the ratio K/L, β-branching $(\beta)^{a}$ and the value of M^2 for the $M4-$ isomers of Sn, Te, Xe, Ba, and Ce.

A \ Z	50^{Sn}	52^{Te}	54^{Xe}	56^{Ba}	58^{Ce}
139					0.740 ± 0.005 [14] 55 ± 3 s [14] 5.8^{c} — 3.6 ± 0.5
137				0.6616 ± 0.0001 2.60 ± 0.11 m 5.5 — 3.6 ± 0.1	0.255 ± 0.002 34.5 ± 0.5 h [15] 2.3 — 2.9 ± 0.4
135			0.51 ± 0.01 14.5 ± 2.0 m 5.0^{c} — 6.0 ± 1.8	0.269 ± 0.002 28.7 ± 0.5 h 2.0^{b} — 2.8 ± 0.3	
133		0.33 ± 0.05 63 ± 7 m 4^{c} — 40	0.2328 ± 0.0003 2.2 ± 0.1 d 2.9 — 4.0 ± 0.3	0.276 ± 0.002 38.8 ± 0.5 h 3.2 — 2.2 ± 0.3	
131		0.180 ± 0.002 30 ± 1 h 2.0 79 4.6 ± 0.6	0.1636 ± 0.0008 12.0 ± 0.6 d 2.3 — 3.6 ± 0.3		

129		0.1060 ± 0.0002 37 ± 5 d [12] 1.0 — [12] 6.1 ± 0.9	0.1966 ± 0.0006 8.0 ± 0.2 d 2.1 — 2.4 ± 0.2		
127		0.0885 ± 0.0004 103 ± 13 d 0.75 2 [13] 4.3 ± 0.5			
125		0.1097 ± 0.0004 62.5 ± 4.5 d 1.1 — 3.8 ± 0.4			
123		0.0885 ± 0.0004 112.6 ± 8.5 d 0.75 — 4.3 ± 0.4		E_γ (Mev) $T_{\frac{1}{2}}$ K/L β (%) M^2	
121		0.0820 ± 0.0004 140 ± 15 d 0.75 — 5.2 ± 0.6			
119	0.065 ± 0.001 247 ± 13 d 0.5 — 7.2 ± 1.5				
117	0.156 ± 0.001 14.5 ± 0.5 d 2.2 — 5.7 ± 0.6				

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[a] Where no value is given the branching is supposed to be < 2 %.

[b] If the theoretical K/L ratio (3.0) is used the value of M^2 is 3.3 ± 0.3.

[c] Theoretical value.

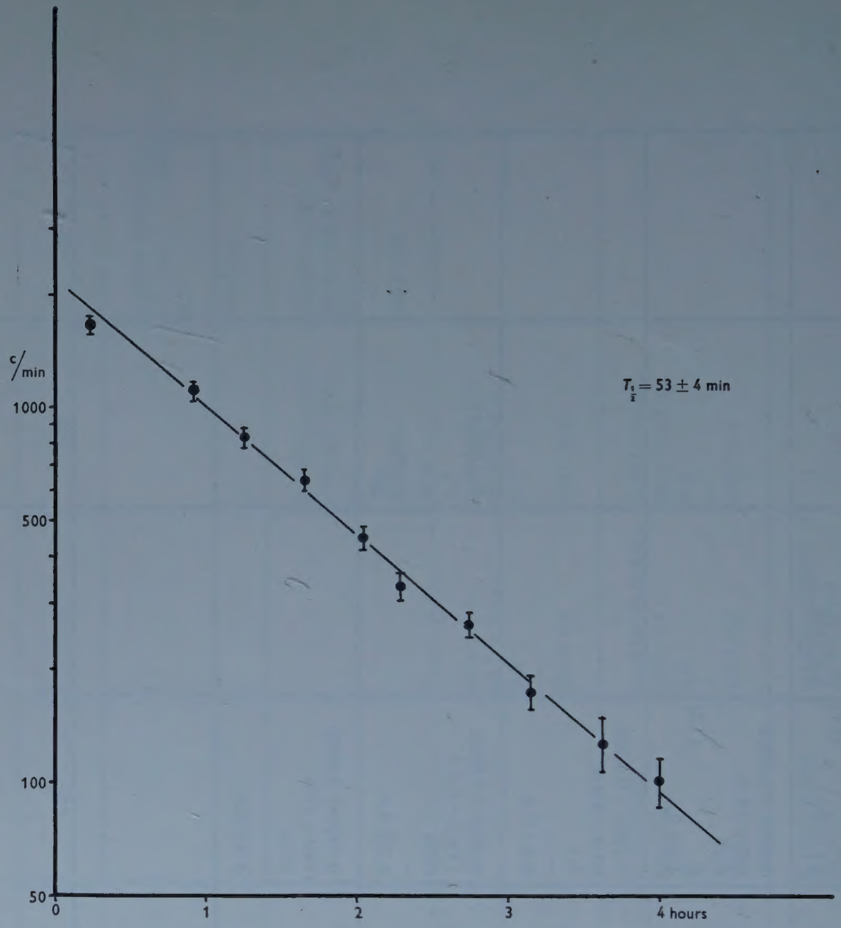

Fig. 2. Half-life of the *K*-conversion line of Te^{133m} measured in a double-focusing spectrometer.

Source preparation.

Uranyl nitrate ($UO_2(NO_3)_2 \cdot 6H_2O$) was irradiated in the reactor of AB Atomenergi, Stockholm, for about 1 hr (flux $= 3 \times 10^{11}$ neutrons/cm^2/sec). The chemical separation followed two different lines.

(A) For the measurements made in the double focusing spectrometer, a carrier-free source of the active tellurium isotopes was required.

The carrier-free tellurium separation was based on the method described in [17]. The irradiated $UO_2(NO_3)_2 \cdot 6H_2O$ was dissolved in water and fumed to near dryness twice with HBr to remove Se, I, Xe, Sb. Inactive Se carrier was added and precipitated with SO_2 from 3 *N* HCl solution, carrying with it the active Te. After repeating this precipitation, the Se carrier was separated from the Te by precipitation from ice-cooled concentrated HCl. A further fuming with HBr removed the remaining traces of Se. A last fuming with concentrated HCl was made to remove the HBr and leave

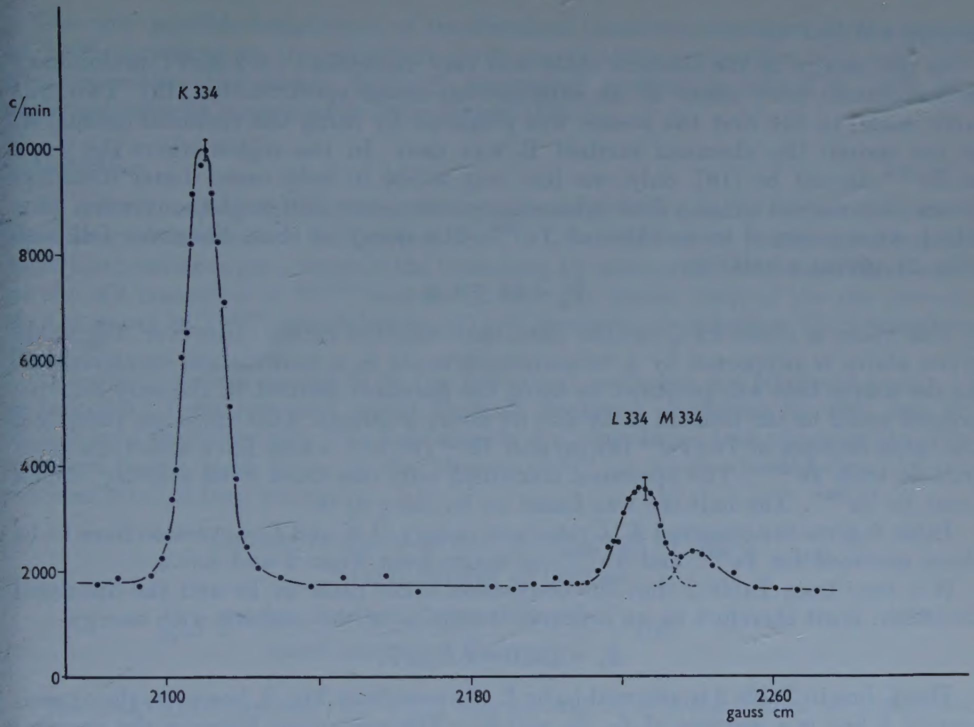

Fig. 3. Conversion line of the isomeric γ-ray of Te^{133m} measured with a resolution 0.5 %.

the final product as the chloride, which is more convenient for the preparation of the sources. Carrier-free Te activity was taken up in approximately 15 cc H_2O.

(B) For certain measurements the chemical isolation of the metastable isomer Te^{133m} was desirable. The method used is taken from Pappas [18] and is based on the fact that a reduction from the oxidation state (VI) to (IV) accompanies the isomeric transition from Te^{133m} to Te^{133}. The separation requires a few mg of Te carrier. The essential separation step freeing the Te (VI) from the growing in Te (IV) is the precipitation of Te (IV) from the cold telluric acid solution with H_2S. The supernatant telluric acid containing pure Te^{133m} is then reduced with HBr and also precipitated with H_2S.

Attempts were made to produce spectrometer sources by evaporating the tellurium activity directly onto a metal foil of the required size, forming the source backing. However, this technique proved to be too time-consuming and, due to the difficulty of evaporating the tellurium chloride, sufficiently strong sources could not be made.

A simple electrolysis technique for making the required spectrometer sources was found to be the most convenient in this case. The final aqueous, carrier-free tellurium cloride solution was electrolyzed between a platinum anode and a foil cathode. This cathode was a 5 mg/cm² thick Cu-foil, painted with nail-polish to leave only the required size of the metal free for the electrodeposition of the tellurium activity. Approximately 0.5 mg NH_2NH_2HCl was added to the solution in order to increase the current to 0.5 A (15 V). A yield of about 40% of the separated activity was obtained on the cathode after electrolysing for 10 minutes.

Energy and half-life

As the energy of the isomeric state was very uncertain (~ 0.3 MeV), preliminary measurements were made in an intermediate-image spectrometer [19]. Two runs were made; in the first the source was prepared by using the chemical method A, in the second the chemical method B was used. In the region where the γ-ray of Te^{133m} should be [18], only one line was found in both cases. Later measurements were carried out in a double focusing spectrometer [20] on the conversion lines which were assumed to be those of Te^{133m}. The decay of these lines was followed (Fig. 2), giving a half-life:

$$T_{\frac{1}{2}} = 53 \pm 4\,\text{m}.$$

This value is about 15% smaller than that reported earlier. However, the value given above is supported by a measurement made in a scintillation spectrometer. As the source here was prepared by using the chemical method B, the only activity present could be the isomers of Te and its decay products. This excludes, therefore, the other isotopes of Te (Te^{134} (44 m) and Te^{129} (70 m)), which have a half-life comparable with Te^{133m}. The spectrum contained only one short lived activity, which must be Te^{133m}. The half-life was found to be (55 ± 6) m.

Table 2 gives the observed K/L ratio and energy of K and L conversion lines to be those assumed for Te^{133m} and Te^{131m}, as taken from Figs. 3 and 4.

It is seen from Table 2 that the conversion takes place in Te and the discussed transition must therefore be an isomeric transition of the element with energy

$$E_\gamma = 334.0 \pm 0.3 \text{ keV}.$$

The L line in Table 2 is assumed to be L_I. As seen from Fig. 3, however, the experimental L line is a mixture of L_I, L_II and L_III. The agreement between the energies of K and L lines in the case of conversion in Te is therefore better than given in Table 2.

For a transition of this half-life and energy the calculations of Moszkowski [2] and the empirical analysis of Goldhaber and Sunyar [21] indicate that the only possible assignments are $M4$ or $E4$. The conversion ratios K/L for a 334 keV transition having either of these multipolarities are found from the tables of Rose [10].

$M4$	4.1
$E4$	2.1

The value obtained for $K/L = 4.5 \pm 0.6$ indicates therefore an $M4$ transition.

Table 2. K/L ratio and energy of the conversion lines of the isomeric γ-ray in Te^{133m} and Te^{131m}.

Assumed isomer	K/L	Measured energy (keV)	Energy of the γ-ray if conversion takes place in		
			Sb	Te	I
Te^{133m}	4.5 ± 0.6	K 302.2 ± 0.3 L 329.3 ± 0.3	332.7 ± 0.3 334.0 ± 0.3	334.0 ± 0.3 334.3 ± 0.3	335.4 ± 0.3 334.5 ± 0.3
Te^{131m}	2.3 ± 0.4	K 149.9 ± 0.2 L 177.1 ± 0.2	180.4 ± 0.2 181.0 ± 0.2	181.7 ± 0.2 182.1 ± 0.2	183.1 ± 0.2 182.3 ± 0.2

The only possible assignment of the discussed transition appears to be the isomer of Te^{133}, according to the argument given above and a comparison with older (18) measurements. One cannot, however, completely exclude an isomer in Te^{134}, but such assignments appear to be very improbable.

β-branching

To determine β-branching from an isomeric state, it is usually necessary to know the decay of the isomeric and ground state in detail. However, in the case discussed here, it is possible to get a value of the branching by comparing the measured intensity of the $M4$ transition in Te^{133m} and Te^{131m} with the fission yield of the two isomers. As the decay of Te^{131m} is well known (11), it is possible to calculate the β-branching in Te^{133m}.

Tellurium isomers (called B in the following) in fission are produced partly by independent formation and partly through decay of precursors (in this case Sb, called A in the following). The fission yelds are, therefore, given either as the independent fission yield (y_i) or as the "fission yield" (y), where also the decay of precursors is taken into account.

$$y = \frac{\text{number of atoms formed directly and by decay}}{\text{total number of fissions}} \times 100,$$

$$y_i = \frac{\text{number of atoms formed directly}}{\text{total number of fissions}} \times 100.$$

The total disintegration rate D_B of the amount of B present after an irradiation time T and a decay time t can be written (cf. e.g. Pappas [18])

$$D_B = R \frac{y_A}{\lambda_A - \lambda_B} \{\lambda_A (1 - e^{-\lambda_B T}) e^{-\lambda_B t} - \lambda_B (1 - e^{-\lambda_A T}) e^{-\lambda_A t}\} + R (y_i)_B (1 - e^{-\lambda_B T}) e^{\lambda_B t},$$

where R is the neutron flux times the amount of material (Uranylnitrate) irradiated, and λ_A; λ_B = the distintegration constants of A and B, respectively.

If the time t is large compared to the half-life of A, this nuclide will have decayed away and, if $\lambda_A \gg \lambda_B$, the expression for D_B above can be written

$$D_B = R\, y_B (1 - e^{-\lambda_B T}) e^{-\lambda_B t}. \tag{2}$$

In the present case

$$Sb^{133}\ (4\ \text{m}) \rightarrow Te^{133m}\ (53\ \text{m})$$

$$Sb^{131}\ (23\ \text{m}) \rightarrow Te^{131m}\ (30\ \text{h})$$

which approximately fulfil the condition $\lambda_A \gg \lambda_B$.

Table 3.

Isomer	T	t	y_B	$T_{\frac{1}{2}}$	β
Te^{131m}	1 h	2.8 h	0.43 %	30 h	79 %
Te^{133m}	1 h	2.8 h	4.60 %	53 m	

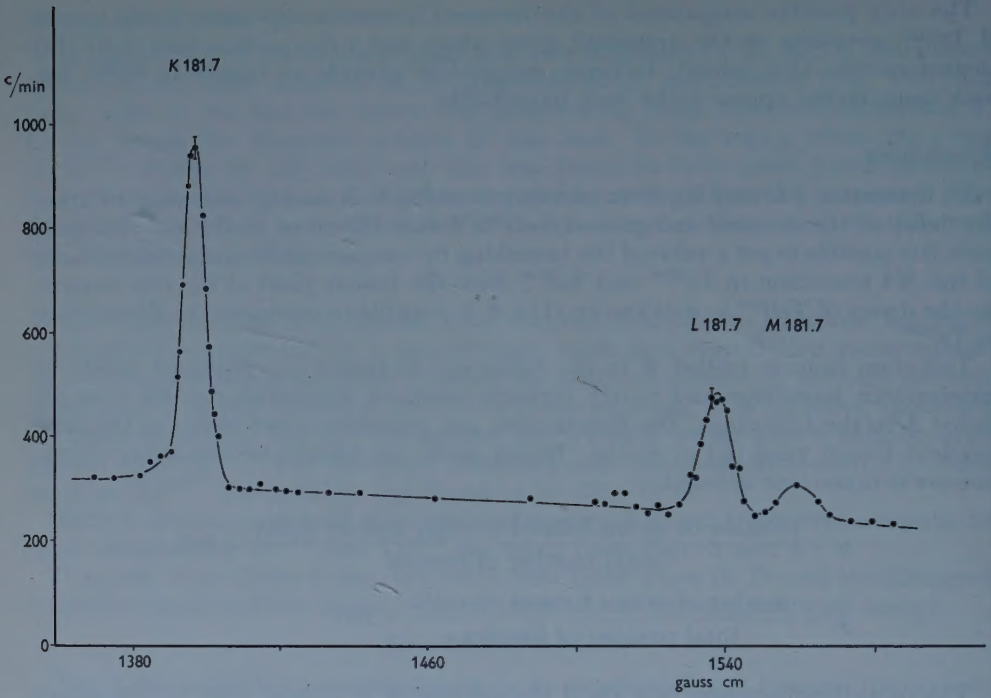

Fig. 4. Conversion lines of the isomeric γ-ray of Te^{131m}. Resolution 0.5 % and corrected to the same time as the measurement of Te^{133m} in Fig. 3.

Table 3 gives the values for one run. The yields are mean values from different authors [17, 18] differing in a few per cent only. The yields are in principle measured from the activity of the Te-daughters I, and the possible neglection of β-branching in those measurements will influence the yield very little because of the short half-lives of the ground state of the Te isotopes in comparison with those of the isomeric states.[1]

From (2) one now finds

$$\frac{D_{Te^{131m}}}{D_{Te^{133m}}} = 0.033. \tag{3}$$

The uncertainty in this figure is about 10%, coming from errors in the yield and half-life and the approximative nature of (2). The latter uncertainty is due to the fact that the time between the end of the irradiation and that time at which Sb is separated away from Te cannot be too long because of the short half-life of Te^{133m}.

The disintegration rates can be measured in a β-spectrometer. At a given time t, D_B can be written

$$D_B = c \cdot I_K \frac{1+\alpha}{\alpha_K} \cdot \frac{100}{100-\beta}, \tag{4}$$

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[1] This assumption has been confirmed by Dr. Pappas in a private communication.

where c = the transmission in the spectrometer and I_K = the intensity of the K-line to be measured.

Fig. 4 gives the spectrum of Te^{131m} measured in the same run as Te^{133m} (Fig. 3) and corrected to the same time. That the conversion took place in tellurium was verified from the energy difference between the K and L lines (Table 2). The ratio $K/L = 2.3$ is in agreement with older measurements and indicates that the transition is of the $M4$ type. The energy is found to be $E_\gamma = 181.7 \pm 0.2$ keV. By comparing the area under the two K-lines in Figs. 3 and 4 one finds from (4)

$$\frac{D_{\mathrm{Te}^{131m}}}{D_{\mathrm{Te}^{133m}}} = 0.0025\,(100 - \beta_{\mathrm{Te}^{133m}}).$$

From (3) β can be calculated to be $\beta_{\mathrm{Te}^{133m}} = 87$. The magnitudes giving rise to uncertainty in (4) are the measured K intensities and the ratio K/L. The error can be estimated to 15%. Adding this error and that from (3) one finds for the branching in Te^{133m}

$$\beta = (87 \pm 3)\%.$$

Thus the isomeric level in Te^{133m} decays not only by the emission of γ-rays and conversion electrons, but also by a β-branch which is nearly 90%.

Discussion

From the measured energy, half-life and β-branching, it is now possible to calculate M^2 of Te^{133m}:

$$M^2 = 5.4 \pm 1.5.$$

Thus, because of the large β-branching, the empirical value of M^2 for Te^{133m} is brought down to the same level as the matrix elements of the other Te isomers. These do not show any marked tendency, now, to increase with the neutron number. There is, however, a small scattering in the values of M^2 and, especially for Te^{133}, the uncertainty is large. The β-branching of this isomer has to be checked by a more direct method. The difficulties here are, of course, the short half-life of the ground state of Te^{133} and the β-decaying, 44-minute Te^{134} formed in high yield in fission. It is possible chemically to separate away the Te^{134}, but as carrier of mg quantities must be used, it is not possible to prepare thin sources for measurements of β-intensities in a beta spectrometer. The only way to get pure Te^{133m} sources appears to be by the use of an electromagnetic isotope separator.

The empirical M^2 values of the Xe and Ba isomers show, as pointed out earlier, a small increase with neutron number, even when the experimental errors are taken into account. There are, however, especially in the Xe group, insufficient measurements yet to be sure of the pronounced effect. It is the last Xe isomer Xe^{135m}, which shows the largest uncertainty caused by the lack of an accurate energy estimate. Finally, in the case of Sn and Ce there are still too few known isomers to give any indication of a corresponding tendency.

ACKNOWLEDGEMENTS

One of us (G. O.) whishes to thank Professor Manne Siegbahn, Head of the Nobel Institute for Physics, for the hospitality extended to her during her stay in Sweden. Our thanks are due to Dr. I. Bergström for pointing out the problem and for many valuable discussions. We wish also to thank Dr. Pappas for kindly making some interesting comments. Finally we thank the reactor group of AB Atomenergi for the irradiations.

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Tryckt den 31 juli 1957

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